# Quarterly Report on the Ferrocyanide Safety Program for the Period Ending September 30, 1996

R. J. Cash J. E. Meacham

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# QUARTERLY REPORT ON THE FERROCYANIDE SAFETY PROGRAM FOR THE PERIOD ENDING SEPTEMBER 30, 1996

# R. J. Cash J. E. Meacham

### **ABSTRACT**

This is the twenty-second and last quarterly report on the progress of activities addressing the Ferrocyanide Safety Issue associated with Hanford Site high-level radioactive waste tanks. No further reports on the Ferrocyanide Safety Program will be issued because the Defense Nuclear Facilities Safety Board closed Recommendation 90-7 (FR 1990) in September 1996. Furthermore, the U.S. Department of Energy also authorized removal of the remaining 14 ferrocyanide tanks from the Watch List in September. All six parts of DNFSB Recommendation 90-7 have been completed. Ferrocyanide is no longer considered a hazard in the Hanford Site high-level waste tanks; the tanks have been reclassified as safe.

This report lists the final accomplishments of the Ferrocyanide Safety Program listed.

Section 1.2 summarizes the key events occurring this quarter. Chapters 2 through 4 discuss program accomplishments in more detail. The 21 previous issues of this quarterly report contain a history of the Ferrocyanide Safety Program.

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# WHC-EP-0474-22

# **CONTENTS**

1.0	INTRODUCTION	1-1
	1.1 PURPOSE	1-1
	1.2 QUARTERLY HIGHLIGHTS	1-1
	1.3 REPORT FORMAT	1-2
	1.4 BACKGROUND	1-2
2.0	FERROCYANIDE SAFETY DOCUMENTATION	2-1
3.0	ACTIONS TO COMPLETE DNFSB RECOMMENDATION 90-7	3-1
	3.1 ENHANCED TEMPERATURE MEASUREMENT	
	3.1.1 Instrument Trees	
	3.1.2 Upgrades to Existing Temperature Monitoring Instrumentation	
	3.1.3 Hot Spot Thermal Modeling	
	3.1.4 Infrared Scanning System	
	3.1.5 Cooling System Requirements	
	3.2 CONTINUOUS TEMPERATURE MONITORING	
	3.3 COVER GAS MONITORING	
	3.3.1 Interim Flammable Gas Monitoring	
	3.3.2 Continuous Gas Monitoring	
	3.4 FERROCYANIDE WASTE CHARACTERIZATION	
	3.4.1 Ferrocyanide Tank Waste Sampling and Analyses	
	3.4.2 Estimation of Water Content	
	3.4.3 Moisture Retention Properties of Ferrocyanide Sludge and Saltcake	
	Simulants	3-12
	3.5 CHEMICAL REACTION STUDIES	
	3.5.1 Chemical Reaction Studies at Pacific Northwest National Laboratory	
	3.5.2 Preparation and Characterization of Ferrocyanide Simulants	
	3.6 EMERGENCY RESPONSE PLANNING	
	5.0 EMERGENCI RESIGNADE I ELITATION	J 17
4 0	IMPLEMENTATION OF THE WYDEN AMENDMENT	4-1
	4.1 THE WATCH LIST	
	4.2 TEMPERATURE MONITORING	
	4.3 PRESSURE MONITORING	
		-
5.0	PROGRAM SCHEDULES AND MILESTONES	5-1
6.0	REFERENCES	6-1
AP	PENDIX	
<b>A</b> :	FERROCYANIDE TANK INFORMATION SUMMARY	. <b>A</b> -3

# WHC-EP-0474-22

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LIST OF FIGURES					
5-1 Ferrocyanide Waste Tank Safety Schedule	e				

### LIST OF TERMS

BIO Basis for Interim Operation

cal/g calories per gram

DNFSB Defense Nuclear Facilities Safety Board

DOE U.S. Department of Energy

DOE-HO U.S. Department of Energy, Headquarters in Washington, D.C.

DOE-RL U.S. Department of Energy, Richland Operations Office

DQO data quality objectives
EA environmental assessment
EMI electromagnetic induction
FAI Fauske and Associates, Inc.
FSAR Final Safety Analysis Report

FY fiscal year g-mol gram-mole

GAO U.S. General Accounting Office

IR infrared

ISB interim safety basis

kW kilowatt

LANL Los Alamos National Laboratory

LOW liquid observation well

NASA National Aeronautics and Space Administration

NIR near infrared

PNNL Pacific Northwest National Laboratory

ppmv parts per million by volume

SA safety assessment SST single-shell tank TC thermocouple

TCR tank characterization report

TMACS Tank Monitor and Control System

USQ unreviewed safety question

WHC Westinghouse Hanford Company

wt% weight percent

# WHC-EP-0474-22

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### 1.0 INTRODUCTION

### 1.1 PURPOSE

This quarterly report provides a final status of the activities that have been conducted to resolve the Ferrocyanide Safety Issue at the Hanford Site over the past 6 years. Many of the tasks were performed in response to Defense Nuclear Facilities Safety Board (DNFSB) Recommendation 90-7 (FR 1990). In March 1991, a DNFSB implementation plan (Cash 1991) responding to the six parts of Recommendation 90-7 was prepared and sent to the DNFSB. A ferrocyanide safety program plan addressing the total Ferrocyanide Safety Program, including the six parts of DNFSB Recommendation 90-7, was released in October 1994 (DOE 1994b). Activities in the program plan have now been completed, as described in Chapters 2 and 3 of this report.

# 1.2 QUARTERLY HIGHLIGHTS

The following activities were the highlights of the Ferrocyanide Safety Program for the fourth quarter of fiscal year (FY) 1996.

- The technical basis safety document (WHC-SD-WM-SARR-038, Revision 1) for removal of the remaining 14 ferrocyanide tanks from the Wyden Amendment Watch List, and for final resolution of the Ferrocyanide Safety Issue, was completed and submitted to DOE-RL on July 3, 1996 (Bacon 1996c, Meacham et al. 1996).
- Final approval was received (Kinzer 1996b) from the U.S. Department of Energy (DOE), Richland Operations Office (DOE-RL), on September 4, 1996 to remove the remaining 14 ferrocyanide tanks (see Table A-1 in Appendix A) from the Wyden Amendment Watch List (Public Law 101-510, 1990). DOE approval to remove the remaining 14 tanks resolves the Ferrocyanide Safety Issue and allows applicable safety documentation to be updated to show this change. A final letter from DOE-Headquarters (DOE-HQ) has been prepared stating that the safety issue is resolved. Receipt is expected in October 1996.
- The Defense Nuclear Facilities Safety Board closed Recommendation 90-7 on September 4, 1996, which constituted the majority of the work assigned to and completed by the Ferrocyanide Safety Program over the last 6 years.
- Four tank characterization reports (TCRs) were issued this quarter for sampling events that were completed earlier this FY. The reports cover tanks 241-BY-104, BY-105, BY-106, and BY-110.

# 1.3 REPORT FORMAT

Progress reports for activities under each of the six parts of DNFSB Recommendation 90-7 are arranged in the same order as the program plan (DOE 1994b). The arrangement also follows the order of Recommendation 90-7. To report on progress, each part of the recommendation is repeated in italics, followed by paragraphs explaining the scope of work on each part or subpart of the recommendation. Subheadings for each task activity report the following:

- Progress during the reporting period
- Planned work for subsequent months
- Problem areas and action taken
- Milestone status.

### 1.4 BACKGROUND

Since the mid-1940's, various high-level radioactive wastes from defense operations have accumulated at the Hanford Site in underground storage tanks. During the 1950's, additional tank storage space was required to support the defense mission. To obtain this additional storage volume within a short time period, and to minimize the need for constructing additional storage tanks, Hanford Site scientists developed a process to scavenge <sup>137</sup>Cs and <sup>90</sup>Sr from tank waste liquids. In implementing this process, approximately 140 metric tons (154 tons) of ferrocyanide were added to waste that was later routed to a number of Hanford Site single-shell tanks (SSTs) (Sloat 1954, 1955).

In the presence of oxidizing material such as sodium nitrate and/or nitrite, ferrocyanide can be made to react exothermically by heating it to high temperatures or by applying an electrical spark of sufficient energy (Cady 1993). However, fuel, oxidizers, and temperature are all important parameters. If fuel, oxidizers, or high temperatures (initiators) are not present in sufficient amounts, a runaway or propagating reaction cannot occur.

In 1990, little was known about the potential hazards of a ferrocyanide-nitrate/nitrite reaction in Hanford Site SSTs. The existing safety analysis report (Smith 1986) and subsequent analyses, such as the 1987 environmental impact statement (DOE 1987), did not adequately define the conditions necessary to preclude propagating reactions in the ferrocyanide waste. Because the safety envelope was not adequately defined by existing analyses, an inadequacy

existed in the authorization basis\*, and an unreviewed safety question (USQ) was declared (Deaton 1990).

Based on the knowledge gained from simulant studies, theoretical analyses, and analyses of actual waste samples, safety criteria were defined for the ferrocyanide waste (Postma et al. 1994a). These criteria were reviewed and accepted by outside reviewers and reviewers within DOE. The USQ was closed on March 1, 1994, by the DOE Assistant Secretary for Environmental Restoration and Waste Management (Sheridan 1994b).

In September 1990, an ad hoc task force report (Kress et al. 1990) recommended that studies be performed to provide information on the potential for a ferrocyanide-nitrate/nitrite explosion, the conditions necessary in the tanks to initiate an explosion, and the potential consequences of such an occurrence. The U.S. General Accounting Office (GAO) advised the Secretary of Energy to implement these recommendations (Peach 1990). A closeout report addressing all three GAO recommendations was submitted to DOE in June 1994 (Payne 1994b). The closeout report summarizes the progress made on determining the potential for ferrocyanide reactions in Hanford Site ferrocyanide tanks, and the conditions necessary to sustain an exothermic ferrocyanide reaction.

In March 1989, based on process knowledge, process records, transfer records, and log books, 22 Hanford Site tanks were identified as potentially containing 1,000 gram-moles (g-mol)\*\* (211 kg [465 lb]) or more of ferrocyanide as the Fe(CN)<sup>4</sup> anion (Nguyen 1989). Two additional ferrocyanide tanks were identified in January 1991 (Borsheim and Cash 1991), increasing the number of ferrocyanide tanks to 24. To avert possible injury to personnel and damage to the facility or environment, strict controls were identified for these and other safety issue tanks in *Operating Specifications for Watch List Tanks* (WHC 1990). Tanks identified by this document (see WHC 1996 for the latest revision) have been commonly referred to as Watch List tanks. In October 1990 (Deaton 1990), the Ferrocyanide Safety Issue was declared a USQ (see Chapter 2) because the safety envelope for these tanks was no longer considered to be bounded by the existing safety analysis report (Smith 1986).

In November 1990, the Wyden Amendment was enacted (Public Law 101-510, 1990). This law required the identification of Hanford Site tanks that may have a serious potential for release of high-level waste (see Chapter 4). In February 1991, the 24 ferrocyanide tanks

<sup>\*</sup>The DOE authorization basis characterizes the facility design basis and operational requirements for each nuclear facility. The authorization basis is described in documents such as facility safety analysis reports and other safety analyses, hazard classification documents, technical safety requirements, DOE-issued safety evaluation reports, and facility-specific commitments such as safety assessments for specific tank operations and the Interim Safety Basis (ISB) (Wagoner 1993).

<sup>\*\*</sup>The 1,000 g-mol criterion has since been replaced with a 115 calories per gram (cal/g) (480 J/g) fuel concentration criterion (see Section 4.1).

### WHC-EP-0474-22

were among the tanks identified (Harmon 1991), and were included in the subsequent July 1991 report to Congress that responded to the Wyden Amendment (Watkins 1991). However, reexamination of the historical records indicated that 6 of the 24 tanks did not contain the requisite 1,000 g-mol of ferrocyanide (Borsheim and Simpson 1991). Therefore, these six tanks should not have been included on the Watch List nor identified in the response to the Wyden Amendment. The six tanks were subsequently removed from the Watch List (Anttonen 1993, Sheridan 1994b).

# 2.0 FERROCYANIDE SAFETY DOCUMENTATION

The USQ process depends on an authorization basis that describes those aspects of the facility design basis and operational requirements relied on by DOE to authorize operation. The authorization basis is described in documents such as facility safety analysis reports and other safety analyses, hazard classification documents, technical safety requirements, DOE-issued safety evaluation reports, and facility-specific commitments such as safety assessments (SAs), the ISB, the Basis for Interim Operation (BIO), and the Final Safety Analysis Report (FSAR) that is scheduled to DOE-RL in November 1996. The potential hazards of a ferrocyanide-nitrate/nitrite reaction were discovered to represent an inadequacy in the authorization basis (Smith 1986). The Ferrocyanide USQ was closed on March 1, 1994 by the DOE Assistant Secretary for Environmental Restoration and Waste Management (Sheridan 1994a). Progress on the remaining safety documentation for resolving the Ferrocyanide Safety Issue is reviewed in this chapter.

Safety and Environmental Assessments. SAs are documents prepared to provide the technical basis for assessing the safety of a proposed activity and to provide proper controls to maintain safety. The SA and the accompanying environmental assessment (EA) for that operation provide the basis for DOE authorization of the proposed activities. SAs were previously approved for headspace sampling of the former ferrocyanide tanks, waste surface sampling, push-mode and rotary-mode core sampling, thermocouple (TC)/instrument tree installation in sound and assumed-leaker tanks, and removal of pumpable liquid (interim stabilization). A generic EA covering all proposed operations in the tank farms was approved and a Finding of No Significant Impact was issued by DOE (Gerton 1994). Approval of the generic EA provides adequate National Environmental Policy Act coverage for the planned Ferrocyanide Safety Program activities.

The authorization basis for intrusive tank operations was combined into one document, the ISB, which was approved in November 1993 (Wagoner 1993). This safety document was updated to reflect the approved ferrocyanide safety criteria and closure of the Ferrocyanide USQ. The ISB will also be updated during the first quarter of fiscal year (FY) 1997 to incorporate recent approvals to remove all ferrocyanide tanks from the Watch List, as described below. This information was incorporated into the BIO submitted to DOE-RL on September 30, 1996, and also into the draft FSAR that will replace the ISB (and BIO as well) when approved by DOE.

Hazard Assessment. A report assessing the ferrocyanide waste tank hazards was issued in July 1992 (Grigsby et al. 1992). The report reviewed the understanding of the ferrocyanide hazard at that time, and presented an integrated evaluation and interpretation of historical data and then-available information. Additional data are now available on the potential for exothermic ferrocyanide reactions in Hanford Site SSTs.

The ferrocyanide hazard assessment document, Assessment of the Potential for Ferrocyanide Propagating Reaction Accidents, WHC-SD-WM-SARR-038, Revision 0 (Grigsby

et al. 1996a), was transmitted to DOE-RL on January 31, 1996. Minor comments were incorporated into the document as Revision 0A and the document was retransmitted to DOE-RL on March 19, 1996 (Grigsby et al. 1996b). Revision 0A was subsequently forwarded to the DNFSB by DOE-RL on April 10, 1996 (Trine 1996). The document provided the technical basis for removal of the four C-Farm ferrocyanide tanks (241-C-108, C-109, C-111, and C-112) from the Watch List. Approval for their removal from the Watch List was received from DOE-RL on June 25, 1996 (Kinzer 1996).

Revision 1 of the document (Meacham et al. 1996) was completed last quarter (Dukelow et al. 1996), addressing all 18 ferrocyanide tanks, and the document was transmitted to DOE-RL on July 3, 1996. The transmittal letter (Bacon 1996c) requested approval to remove the remaining 14 ferrocyanide tanks from the Watch List and for final resolution of the Ferrocyanide Safety Issue based on Revision 1 of the document. The Chemical Reaction Subpanel of the Tanks Advisory Panel reviewed the document in June 1996, and their comments were addressed and incorporated. Technical information from all Ferrocyanide Safety Program tasks was compiled into this document, and following DOE approval, the ISB and draft FSAR will be revised accordingly.

Approval to remove the remaining 14 ferrocyanide tanks from the Watch List was received from DOE-RL on September 4, 1996 (Kinzer 1996). The DNFSB also closed Recommendation 90-7 on September 4, 1996 (Conway 1996). These actions are reflected in the BIO that was submitted to DOE-RL for approval on September 30, 1996. Changes to the ISB, OSD-T-151-00030 (WHC 1996), and the draft FSAR will be completed during the first quarter of FY 1997.

Ferrocyanide Program Plan. A ferrocyanide program plan was submitted to the DNFSB in December 1994 (O'Leary 1994). The program plan outlined the activities planned to address DNFSB Recommendation 90-7, to meet the Wyden Amendment requirements (Public Law 101-510, 1990), and to remove the remaining ferrocyanide tanks from the Watch List. In the program plan all ferrocyanide activities were scheduled to be completed by the end of FY 1997. However, an increased understanding of radiolytic and chemical degradation (aging) of ferrocyanide and sampling of bounding tanks allowed the program to be completed in FY 1996 with the removal of the 18 ferrocyanide tanks from the Watch List. This action resulted in resolution of the Ferrocyanide Safety Issue much earlier and at a substantially reduced cost. Former ferrocyanide tanks that have not been sampled to date will eventually be sampled as part of other safety issues or DQOs, such as the Organic Safety Project or the Safety Screening, Historical, or Retrieval DQOs (Dukelow et al. 1995, Simpson and McCain 1996).

### Milestone Status

-- January 31, 1996. Westinghouse Hanford Company issues documentation supporting safety issue resolution for the four C-Farm tanks and recommends their removal from the Wyden Amendment Watch List (Public Law 101-510, 1990). The Assessment of the Potential for

Ferrocyanide Propagating Reaction Accidents, WHC-SD-WM-SARR-038, Revision 0, was transmitted to DOE-RL as scheduled (Bacon 1996a, Grigsby et al. 1996a). All four C-Farm tanks were sampled earlier and data interpretation reports have been completed for these tanks.

The report was revised to incorporate informal comments received from DOE-RL and reissued with the same title, as WHC-SD-WM-SARR-038, Revision 0A (Grigsby et al. 1996b). The revised report was transmitted to DOE-RL with a letter requesting that the four C-Farm tanks be removed from the Watch List on March 19, 1996 (Bacon 1996b). The report was subsequently forwarded to the DNFSB Staff on April 10, 1996 (Trine 1996).

- July 31, 1996. Westinghouse Hanford Company receives DOE approval to remove the four C-Farm ferrocyanide tanks from the Watch List. Approval was received from DOE on June 25, 1996, ahead of schedule (Kinzer 1996a).
- July 31, 1996. Westinghouse Hanford Company prepares and submits the final ferrocyanide hazard assessment for DOE approval, providing the technical basis for removing all 18 ferrocyanide tanks from the Watch List, and resolution of the Ferrocyanide Safety Issue. The official due date for this milestone is August 30, 1996, with an enhanced date of July 3, 1996. The final ferrocyanide hazard assessment, WHC-SD-WM-SARR-038, Revision 1 (Meacham et al. 1996), was prepared and submitted for DOE approval on July 3, 1996 (Bacon 1996c).
- September 30, 1997. Westinghouse Hanford Company receives DOE approval for resolution of the Ferrocyanide Safety Issue. DOE-HQ and DOE-RL have approved removal of the remaining 14 tanks (Kinzer 1996b) from the Watch List. At the time approval was given, DOE-HQ stated that final resolution of the Safety Issue would be granted as soon as the DNFSB closed 90-7. The DNFSB completed that action on September 4, 1996 (Conway 1996). The final letter from DOE-HQ stating that the issue has been resolved currently is being prepared.

# WHC-EP-0474-22

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# 3.0 ACTIONS TO COMPLETE DNFSB RECOMMENDATION 90-7

This chapter follows the format of the program plan (DOE 1994b) and describes all work associated with the Ferrocyanide Safety Program. Where applicable, each task activity is described relative to the DNFSB Recommendation (90-7.1 through 90-7.6). The specific part of the recommendation is given, followed by a summary of activities completed to respond to that part of Recommendation 90-7 (if not already closed out). All parts of Recommendation 90-7 are now closed (Conway 1996), and this is the last quarterly report that will be issued on the Ferrocyanide Safety Program.

### 3.1 ENHANCED TEMPERATURE MEASUREMENT

"Immediate steps should be taken to add instrumentation as necessary to the SSTs containing ferrocyanide that will establish whether hot spots exist or may develop in the future in the stored waste. The instrumentation should include, as a minimum, additional thermocouple trees. Trees should be introduced at several radial locations in all tanks containing substantial amounts of ferrocyanide, to measure the temperature as a function of elevation at these radii. The use of infrared techniques to survey the surface of waste in tanks should continue to be investigated as a priority matter, and on the assumption that this method will be found valuable, monitors based on it should be installed now in the ferrocyanide bearing tanks."

### 3.1.1 Instrument Trees

All new instrument trees planned for the former ferrocyanide tanks have been installed; this action was completed in August 1995. Originally, several new temperature measurement instruments were to be installed into each tank. This plan was modified to ensure that at least one instrument tree with replaceable temperature-sensing elements was in each ferrocyanide tank. The new trees have at least two operational temperature-sensing elements in the waste to ensure a true temperature measurement, and one or more elements in the headspace. The number of temperature sensing elements in the new trees is typically six, but some tanks with larger inventories of waste contain up to eight elements.

The decision to put only one new tree into each tank was based on the following information: many of the TC elements in the existing trees were returned to service in FY 1992, and measured temperatures are as expected (Bussell 1992), thermal modeling (McLaren 1994a, 1994b) and an enhanced understanding of waste properties show that formation of hot spots in former ferrocyanide tanks is not credible (Dickinson et al. 1993, Epstein et al. 1994), and new calculations of tank heat content based on tank temperatures show lower values than previous estimates (Crowe et al. 1993, McLaren 1994a, 1994b).

There are two instrument trees in all but three of the former ferrocyanide tanks (241-BY-106, BY-111, and BY-112). The instrument tree in tank 241-BY-106 already had replaceable temperature sensing elements, and tanks 241-BY-111 and BY-112 previously had no operable instrument trees. The instrument trees in the former ferrocyanide tanks are monitored continuously by the Tank Monitor and Control System (TMACS). The older instrument trees are expected eventually to fail in a manner such that they cannot be repaired; they will not be replaced. The highest temperatures recorded in each tank for the quarter ending September 30, 1996 are listed in Appendix A, Table A-1.

• Status. The last of 16 new instrument trees was installed in the 18 former ferrocyanide tanks in August 1995. Thirty-three working instrument trees and a TC element in each liquid observation well (LOW) of tanks 241-BY-111 and BY-112 are continuously monitored by TMACS. All work is complete for this task and DNFSB Recommendation 90-7.1 is closed.

## 3.1.2 Upgrades to Existing Temperature Monitoring Instrumentation

This task determined the operability and accuracy of previously installed TC elements in the original 24 ferrocyanide Watch List tanks. The original and newly installed instrument trees provide temperature measurements for each of the former ferrocyanide tanks.

Field measurements were taken in 1991 on each TC element in the then-existing trees to determine the resistance and voltage across the junction, and across each lead to ground. The exact condition of each TC element was determined by resistance and voltage measurements (Bussell 1992). This work was completed in FY 1991 with 265 TC elements evaluated. Work in FY 1992 focused on repair and recovery of 92 TC elements that were found to be failed or performing marginally. This task was completed in FY 1992 for the Ferrocyanide Safety Program.

• Status. This task is complete for the Ferrocyanide Safety Program.

### 3.1.3 Hot Spot Thermal Modeling

Radioactive materials decaying in Hanford Site waste tanks generate heat. An early concern, raised when the ferrocyanide tanks first became a safety issue, was whether an exothermic excursion and local propagation could occur within the ferrocyanide waste if a sufficient concentration of ferrocyanide and a high enough temperature were present. This task examined the available temperature data from the ferrocyanide tanks to determine the heat load and temperatures as a function of depth and radial location. Sensitivity and parametric analyses were included to determine the magnitude of a hot spot that would have to exist for the waste to reach propagation temperatures.

Heat load analyses and thermal characteristics were completed for all former ferrocyanide tanks in FY 1994 (McLaren 1994a, 1994b). The maximum heat load of any ferrocyanide tank, assuming worst case conditions for soil moisture and thermal conductivity, was below 4.2 kilowatts (kW). Nominal heat loads calculated in 1994 (McLaren 1994a, 1994b) compared very favorably with those calculated independently in 1993 (Crowe et al. 1993). A dryout analysis was also completed and released in FY 1994 (Epstein et al. 1994). The report concluded that ferrocyanide sludge could not dry sufficiently to be chemically reactive during interim storage, either globally or locally. Dryout mechanisms evaluated included global evaporation, removal of liquid by leakage or pumping, boiling as a result of hot spots, and enhanced surface evaporation from hot spots. All activities were completed for this task in FY 1994.

• Status. This task is complete.

# 3.1.4 Infrared Scanning System

Infrared (IR) scanning systems are commercially available from numerous vendors. These systems are sensitive to changes of  $\pm$  0.3 °C or less under ideal conditions, and offer promise for mapping surface temperature profiles in the high-level waste tanks. Thermal modeling performed on ferrocyanide tank 241-BY-104 suggested that if hot spots with temperatures of concern are possible, surface temperature differences might be great enough to be detected by IR mapping (McLaren 1993).

A position paper on the credibility of hot spots and the need for further IR scanning was issued in April 1993 (Dickinson et al. 1993). Further analyses have been performed to assess potential dryout of the ferrocyanide waste (Epstein et al. 1994). These reports examined potential mechanisms for forming hot spots. Analyses indicate that hot spots are not credible in the former ferrocyanide tanks. Based on these analyses, Westinghouse Hanford Company recommended that no further planning be pursued for IR scans to detect hot spots. Work on this Ferrocyanide Safety Program task was concluded at the end of FY 1993.

• Status. This task is complete.

# 3.1.5 Cooling System Requirements

The program plan for resolution of the Ferrocyanide Safety Issue (DOE 1994b) provided actions that would be taken to cool the ferrocyanide tanks if it were found that such cooling was necessary. Several tentative milestones, identified below, were established for use if a cooling system were required. The concern at the time was that increasing temperatures could lead to loss of moisture within the ferrocyanide waste matrix. Immediate emergency actions that would be taken if increased temperatures were to occur are described in the Action Plan for Response to Abnormal Conditions in Hanford Site Radioactive Waste Tanks

Containing Ferrocyanide (WHC-EP-0407, Revision 2 [Fowler 1994]). Types of cooling systems might include, but are not limited to, the following: forced ventilation of the tank, using an existing or new exhauster system, air conditioning the air to the tank, adding humid air or mist, and adding water to the tank.

Based on the historical database, analytical data on samples from the former ferrocyanide tanks, and results from the Pacific Northwest National Laboratory (PNNL) aging test activity, none of the 18 former ferrocyanide tanks contain a high enough concentration of ferrocyanide for a propagating reaction to occur. Because dryout of the waste under the present storage conditions is not credible, a special cooling system for the former ferrocyanide tanks is not considered necessary (Epstein et al. 1994).

• Status. This task is complete.

### 3.2 CONTINUOUS TEMPERATURE MONITORING

"The temperature sensors referred to above [Recommendation 90-7.1] should have continuous recorded readouts and alarms that would signal at a permanently manned location any abnormally high temperatures and any failed temperature instrumentation."

This task provided continuous monitoring of presently installed (and operable) temperature-sensing elements for the former ferrocyanide tanks. New instrument trees were connected to TMACS shortly after they were installed in each tank, resulting in continuous temperature monitoring in the those tanks. All data are collected automatically at the continuously manned Computer Automated Surveillance System Operator Control Station. The monitoring system is independent of the Computer Automated Surveillance System and displays data to an operator on request. Trend data on selected points are available for display in numeric or graphic form.

The TMACS system, which became operational in September 1991, provides alarms for a change in the value of any temperature point. Alarms, if they occur, trigger an audible annunciator and are logged immediately to hard copy. An alarm summary display provides a list of the most recent alarms in order of occurrence. Each alarm can be identified by point and time of occurrence. Operator acknowledgement of the alarm will silence the audible annunciator. Signal conditioning and multiplexing are performed locally at each tank, eliminating the need to transmit low-level signals to the tank farm boundary and reducing cable runs. Electronic noise, extension wire corrosion, and thermal gradients are also reduced.

• Status. Temperatures measured by 33 instrument trees and two TC elements in LOWs in the former ferrocyanide tanks are being monitored continuously by TMACS. This work was completed in August 1995, and DNFSB Recommendation 90-7.2 is closed. The highest temperatures recorded in each of

the tanks for the quarter ending September 30, 1996 are listed in Appendix A, Table A-1. This task is complete.

### 3.3 COVER GAS MONITORING

"Instrumentation should also be installed to monitor the composition of cover gas in the tanks, to establish if flammable gas is present."

## 3.3.1 Interim Flammable Gas Monitoring

Flammable and toxic gas monitoring and analyses in the former ferrocyanide tanks and other Hanford Site waste tanks are continuing. This effort was transferred to the Tank Vapor Monitoring Program, which is coordinating interim gas monitoring of all Hanford Site tanks involved with the tank vapor program. Tank headspaces are measured for flammability using a commercial combustible gas monitor (calibrated with pentane gas), and are monitored for potential toxic gases using an organic vapor monitor and Dräger\* tubes. Headspace characterization of all the Hanford Site high-level waste tanks is continuing using sorbent tubes placed on the end of tubes lowered into the headspace, and SUMMA\*\* canisters that collect gas samples topside. The initial headspace sampling was done in several tank locations (i.e., from two widely separated risers) and at three elevations in the headspace. Reviews of sampling data and modeling (Wood 1992, Claybrook and Wood 1994, Postrna et al. 1994b) indicate that the headspace is well mixed and that sampling from one riser at one elevation is adequate.

• Status. Headspace sampling of all 18 former ferrocyanide tanks as required for this task was completed in May 1995. The results are summarized in Appendix A, Table A-2. Headspace sampling of the former ferrocyanide tanks and other Hanford Site high-level waste tanks will continue on a periodic basis as part of the Tank Vapor Monitoring Program. DNFSB Recommendation 90-7.3 is closed.

# 3.3.2 Continuous Gas Monitoring

The possibility that localized concentrations or stratification of gases exist in the tanks was evaluated. A modeling study was conducted to determine airflow patterns in the headspace of tank 241-C-109, and to evaluate the amount of mixing and the local gas concentrations that could occur. Since the study revealed that the gases in the tank are well mixed and

<sup>\*</sup>Trademark of Drägerwerk Aktiengesellschaft, Inc., Lubeck, Germany; also National Draeger, Inc., Pittsburgh, Pennsylvania.

Trademark of Molectrics, Inc., Cleveland, Ohio.

follow Graham's law for gaseous diffusion, an analysis of a second tank was considered unnecessary (Wood 1992). Studies completed since that time confirm that conclusion (Claybrook and Wood 1994, Postma et al. 1994b).

The need for continuous gas monitoring was addressed in a report that also assessed the potential for cyclic venting and the possibility of accumulating flammable gases (Fowler and Graves 1994). The report concluded that continuous flammable gas monitoring in ferrocyanide tanks was not warranted based on the low concentration of flammable gases found to date, anticipated low ferrocyanide concentrations because of waste aging, analytical results from tanks 241-C-109 and C-112 showing that the fuel concentration in the tanks is much lower than postulated by flowsheet values and operating records, and calculations of hydrogen accumulation using realistic generation values and passive ventilation assumptions. Vapor sampling of all 18 former ferrocyanide tanks has corroborated that flammable gas concentrations in these tanks are too low to be of concern. No further activities are planned for this task.

• Progress During Reporting Period. This task is complete. DOE concurred that no continuous gas monitoring is required (O'Leary 1994).

# 3.4 FERROCYANIDE WASTE CHARACTERIZATION

"The program of sampling the contents of these tanks should be greatly accelerated. The proposed schedule whereby analysis of two core samples from each single-shell tank is to be completed by September 1998 is seriously inadequate in light of the uncertainties as to safety of these tanks. Furthermore, additional samples are required at several radii and at a range of elevations for the tanks containing substantial amounts of ferrocyanide."

Characterization of the waste in the former ferrocyanide tanks was necessary to guide further chemical reaction studies with the ferrocyanide waste simulants, determine actual chemical and physical properties of the waste, determine how the ferrocyanide waste can be safely stored until retrieval and disposal actions are completed, and apply the study results to the final remediation (exhumation and vitrification) of the waste. This information was used to resolve the Ferrocyanide Safety Issue.

The important reactive materials that were thought to be present in the former ferrocyanide tanks are fuel (ferrocyanides, sulfides, and reduced carbon species such as organic complexants), oxidants (nitrates and nitrites), and inerts or diluents (including phosphates, aluminates, sulfates, carbonates, oxides, hydroxides, and most importantly water). The location of fission products such as <sup>137</sup>Cs and <sup>90</sup>Sr is important because these products are heat sources and potential source terms in postulated radiological releases from a hypothetical ferrocyanide reaction. The water content of the waste is very important because the high heat capacity and heat of vaporization of water make it an effective inerting material. Water can prevent a sustained combustion or a propagating reaction if sufficient fuel is present; wet ferrocyanide material would require drying before it could react or propagate.

# 3.4.1 Ferrocyanide Tank Waste Sampling and Analyses

Tank Sampling. Rotary-mode and push-mode sampling capabilities, auger surface sampling, and grab sampling (primarily for liquids) are used to obtain waste samples from the high-level waste tanks. Tanks without saltcake and with relatively soft waste solids can be sampled by the push-mode method. If a hard saltcake or sludge layer is present, rotary-mode sampling is used. Auger sampling may also be used if the depth of waste is nominally less than 60 cm. Grab sampling is used for tanks containing supernatant and soft sludge waste and was not employed in the former ferrocyanide tanks.

Each core consists of several 48-cm segments (or portions thereof) depending on the depth of the waste in the tank. The sludge layer for cores obtained from the former ferrocyanide tanks was normally divided into four 12-cm subsegments if a full 48-cm segment was obtained; otherwise the sample was subdivided into one or more subsegments that were at least 12 cm in length. If the tank contained a saltcake layer, the saltcake segments were divided into only two subsegments. Segments that contained both sludge and saltcake were divided such that the two types of waste were separated into separate samples. Process flowsheet knowledge, tank historical data, and results obtained from tests with ferrocyanide sludge simulants were used to supplement the analytical results from core sampling.

The priority for sampling former ferrocyanide tanks was changed to reflect the need to determine the reactive properties of the contents. In response to DNFSB Recommendation 93-5 to expedite sampling and analyses required to address safety issues in the Hanford Site Watch List tanks (DOE 1994a), the analysis plans for future ferrocyanide tank core samples (and the plans for other Watch List tanks) were revised in 1993. The Watch List tanks were given priority for core sampling, and the number of required analytes was reduced. Analyte selection was refocused primarily on safety-related properties.

• Progress During Reporting Period. Analytical analyses reports were completed this quarter or during the previous quarter for former ferrocyanide tanks 241-BY-104, BY-105, BY-106, and BY-110. The final laboratory reports have also been issued. Samples obtained did not always meet the requirements specified in the Ferrocyanide DQO document (Meacham et al. 1995), because only one core or parts of one core were obtained. Samples from the attempts to perform push-mode core sampling of tanks 241-BY-105 (riser 12A/core 108) and BY-106 (riser 5/core 121) were analyzed even though the sampling event did not meet the Ferrocyanide DQO requirements. These two tanks were sampled during the first quarter of FY 1996 but no sludge segments were recovered because the downforce limit was reached and sampling was stopped.

Tank characterization reports were issued for all four of the BY tanks during this past quarter, as shown below:

BY-104: WHC-SD-WM-ER-608, Rev. 0, (Benar et al. 1996) was issued September 26, 1996.

BY-105: WHC-SD-WM-ER-598, Rev. 0, (Simpson et al. 1996a) was issued August 30, 1996.

BY-106: WHC-SD-WM-ER-616, Rev. 0, (Bell et al. 1996) was issued September 26, 1996.

BY-110: WHC-SD-WM-ER-591, Rev. 0, (Simpson et al. 1996b) was issued September 16, 1996.

- Planned Work For Subsequent Months. There are no plans to apply the Ferrocyanide DQO (Meacham et al. 1995) to the unsampled former ferrocyanide tanks formerly on the Watch List; however some of these tanks will be sampled for other issues, as mentioned in Chapter 2. The ferrocyanide hazard assessment document has presented the case that further sampling of additional tanks formerly on the Ferrocyanide Watch List is not necessary, and DOE-HQ and DOE-RL have agreed and approved removal of all ferrocyanide tanks from the Watch List.
- Problem Areas and Actions Taken. None required.
- Milestone Status.
  - December 31, 1995. Westinghouse Hanford Company obtains core samples from five additional ferrocyanide tanks. Only one ferrocyanide tank, 241-BY-104, was successfully sampled in November 1995. Attempts to obtain push-mode core samples from tanks 241-BY-105 and BY-106 were discontinued when the downforce limit was reached for the drill string. There are no plans to sample additional tanks for ferrocyanide purposes. This milestone was superseded by the final hazard assessment document (WHC-SD-WM-SARR-038, Revision 1), which was issued July 3, 1996 (Meacham et al. 1996).
  - March 31, 1996. Westinghouse Hanford Company completes data interpretation reports, available for public release, for five ferrocyanide tanks. This milestone date was not met because of the delays encountered in rotary-mode sampling of the tanks. Because the 18 ferrocyanide tanks have been removed from the Watch List and DNFSB Recommendation 90-7 has been closed, this milestone is no longer required. TCRs were completed for the four ferrocyanide tanks most recently sampled, 241-BY-104, BY-105, BY-106, and BY-110.
  - July 31, 1996. Westinghouse Hanford Company obtains core samples from the remaining ferrocyanide tanks. This milestone is no longer required because the 18 tanks have been removed from the Watch List and DNFSB Recommendation 90-7 has been closed.

October 31, 1996. Westinghouse Hanford Company completes data interpretation reports (TCRs), available for public release, for the remaining ferrocyanide tanks. This milestone is no longer required.

Infrared Spectroscopy Analyses. Near-infrared (NIR) spectra were collected from archived waste tank core samples with various chemical matrices using Fourier transform infrared spectrometry-based fiber optics. The task was completed in FY 1995.

Final reports summarizing IR work were issued at the end of FY 1995 (Rebagay et al. 1995, Reich et al. 1995, Douglas and Reich 1995).

• Status. This task was completed in FY 1995.

Mössbauer Spectroscopy. A small task on Mössbauer spectroscopy, previously supported by the Ferrocyanide Safety Program, investigated the physical and chemical nature of iron in tank waste. Iron was a major constituent of the original ferrocyanide waste, and information about its location and composition in the tanks might have proved useful for safe interim storage and eventual retrieval of the waste if the program had continued. The Mössbauer program represented a cooperative venture between Westinghouse Hanford Company, DOE, and the National Aeronautics and Space Administration (NASA), which had developed a Mössbauer spectrometer small enough to perform elevation scans within the LOWs of the Hanford Site waste tanks. The contact at NASA was Dr. Richard Morris at the Johnson Space Center in Houston, Texas.

• Status. Funding from the Ferrocyanide Safety Program for this task was terminated in January 1996 when a financial change request was approved by Westinghouse Hanford Company and DOE-RL. The task was deleted from the scope of the Ferrocyanide Safety Program because completion of the effort was not required to resolve the Ferrocyanide Safety Issue.

Scanning Electron Microscopy. Scanning electron microscopy was a small task that investigated the chemical and physical properties of waste stored in the Hanford Site high-level waste tanks. Studies to further refine what is known about the chemical and physical properties of ferrocyanide tank waste could continue if they were necessary for resolving the Ferrocyanide Safety Issue. However, it has been shown with confidence that the ferrocyanide concentration in the tanks has significantly degraded (aged) to the point where the fuel value is much too low to support a propagating reaction. Thus, further work on this task is not necessary.

• Status. Funding from the Ferrocyanide Safety Program for this task was terminated in January 1996 when a financial change request was approved by WHC and DOE-RL. The task was deleted from the scope of the Ferrocyanide Safety Program because completion of the effort was not required to resolve the Ferrocyanide Safety Issue.

### 3.4.2 Estimation of Water Content

Methods for determining water concentrations in Hanford Site waste tanks are being developed using sample data analyses and state-of-the-art surveillance systems. This work was originally sponsored by the Ferrocyanide Safety Program, but it was transferred to the Organic Safety Project at the beginning of FY 1996. Two in situ moisture monitoring technologies are currently under development: neutron diffusion and electromagnetic induction (EMI). Initial development of NIR spectroscopy for determining the water content of Hanford Site tank waste was completed in FY 1994 at the University of Washington Center for Process Analytical Chemistry (Reich et al. 1994), and by Westinghouse Hanford Company in FY 1995 (Rebagay et al. 1995, Reich et al. 1995). A report examining moisture-monitoring technologies was completed in April 1993 (Meacham et al. 1993).

The water content of reactive waste can be extremely important in ensuring the safety of the Hanford Site tank waste. The water concentration within the waste will determine whether or not it will react and propagate. If sufficient water is present, a chemical reaction cannot start. Recent work by Fauske and Associates, Inc. (FAI) has shown that as little as 5 weight percent (wt%) unbound water in a stoichiometric reactive mixture will prevent the mixture from igniting and burning when a significant ignition source is applied (Fauske 1996). Water may become the prime safety factor for the organic waste tanks to ensure that the waste is stored safely. For the former ferrocyanide tanks, it was shown that the present ferrocyanide content is insufficient for the waste to propagate, so the presence of water becomes a second safety barrier ensuring that the tanks are safe.

Neutron Diffusion. Well-logging techniques, coupled with computer modeling, were developed and applied to an existing neutron probe to determine information about the water concentrations, material interfaces, and other waste characteristics in the former ferrocyanide tanks. Using the knowledge gained from computer modeling, in situ measurements, and experimental calibration data with the current in-tank liquid-level neutron probe (Watson 1993), prototype moisture-measurement neutron probes were developed. This system consists of three neutron probes: a near-field thermal neutron probe, a far-field thermal neutron probe, and a far-field epithermal neutron probe. This improved system would primarily be used to determine the axial moisture concentration profile of the waste inside the ferrocyanide tanks.

Moisture measurement using neutron diffusion is an established technology. The technique uses a neutron source and one or more neutron detectors. The thermal neutrons reaching a detector originate as fast neutrons from the source and are slowed or absorbed by the medium. Because hydrogen atoms are effective at slowing down neutrons, the detector response is a strong function of the surrounding moisture concentration.

When using neutron diffusion, two methods are generally used to measure moisture concentration around LOWs. The first method, the moisture gauge, has a short source-to-detector spacing, on the order of 0 to 10 cm (near-field). The response of a moisture gauge is characterized by an increase in detector response with increasing moisture

concentration of the surrounding medium. The second method, the neutron log, often has two detectors with longer source-to-detector spacings, on the order of 20 to 50 cm (far-field). The detectors in a neutron log arrangement exhibit a decreased response to increased moisture concentrations. The detector placed at the shorter spacing is used to correct the response of the longer-spaced detector for borehole effects.

Tank moisture measurements are taken from within LOWs. The LOWs are permanently installed sealed pipes that extend from the riser top through the tank waste to near the tank bottom. The LOWs allow axial information about the surrounding waste materials to be obtained using certain detectors.

The initial design and prototype tests were completed for a new surface-moisture measurement neutron probe in FY 1995. Results and progress on this task were reported in Watson (1993), Finfrock et al. (1994), and Lipke (1995). This effort was transferred to the Organic Safety Project because it was not required by the Ferrocyanide Safety Program to resolve the Safety Issue.

• Status. This task was transferred to the Organic Safety Project at the end of FY 1995.

Electromagnetic Induction Probe. This task deployed the EMI probe in the LOWs or on top of the waste surface, to measure moisture concentration. EMI probes operate by creating a magnetic field that induces current in a conductive medium. This induced current can be measured and is related to the media conductivity. The higher the electrical conductivity, the higher the free moisture content in the tank waste.

The EMI probe is designed with four separate coils of wire that can be either exciting coils or sensing coils. The present configuration uses one coil as the exciting coil and three coils as the sensing coils. This configuration allows three different depths of penetration during one scan. The electronics can be programmed to use four frequencies during one scan, so the information acquired will be four frequencies at three different coil spacings. This information will be useful in separating the environment near the LOW from the environment far from the LOW. Two different EMI probes have been built with different coil spacing and turns per coil to determine in-tank responses.

EMI probes were deployed in tanks 241-BY-104, BY-106, BY-107, BY-111, BY-112, TY-103, TX-118, TX-114 (3 LOWs), S-105, and S-106. Observations of the data acquired by the probes allowed some conclusions about EMI to be made: the system is sensitive to loss of hydraulic conductivity, which occurs at approximately 0.08 to 0.12 volume fraction of liquid, depending on porosity; the scan can interrogate multiple depths simultaneously; EMI is sensitive to small changes in material properties; the EMI method measures conductivity directly, while moisture interpretation requires some assumptions; EMI results are affected by temperature, so compensation is required; and the EMI method is strongly affected by ferromagnetic items. Reports have been issued on this work (Crowe and

Wittekind 1995, Wittekind and Crowe 1996). The remaining development effort was transferred to the Organic Safety Project at the end of FY 1995.

• Status. This task was transferred to the Organic Safety Project at the end of FY 1995.

# 3.4.3 Moisture Retention Properties of Ferrocyanide Sludge and Saltcake Simulants

The water content of reactive sludge, if sufficient, will prevent exothermic fuel/nitrate-nitrite reactions. Studies were completed to evaluate the water retention properties of ferrocyanide tank sludge and saltcake simulants as they relate to possible waste tank leaks, tank stabilization by pumping, and possible evaporation from exposed surfaces. Previous work has shown that ferrocyanide sludge cannot dry sufficiently to be chemically reactive (assuming sufficient fuel is present) during interim storage, either globally or locally (Epstein et al. 1994). Dryout mechanisms evaluated included global evaporation, removal of liquid by leakage or pumping, boiling as a result of hot spots, and enhanced surface evaporation from hot spots. Recent work focused on water retention in saltcake material, especially after a tank has been interim-stabilized. This work was supported by the Ferrocyanide Safety Program until the end of FY 1995, but was transferred to the Organic Safety Project at the beginning of FY 1996.

Modeling calculations were performed to estimate the water-retaining capability of ferrocyanide waste in typical Hanford Site tank systems. The effort focused on evaluating the impact of consolidation and surface evaporation processes. Computer models were used to estimate retention of water within the matrix and to determine surface drying of sludge and saltcake waste. To accomplish these objectives, the hydraulic properties of actual sludges and saltcake porous media were compared with tested waste simulants, and their physical properties were correlated.

Modeling was also performed to examine the resistance of saltcake waste to gravity drainage and surface evaporation. Under the influence of gravity, saturated saltcake will drain when liquid is pumped out and when a tank is stabilized. In contrast, sludge does not readily drain and the interstitial liquid must be expelled by consolidation, usually caused by an overburden. Because saltcake drains when stabilized, it is more subject to potential drying at the surface as a result of water evaporation.

• Status. Modeling of the water retention properties of saltcake and sludge waste was completed and documented in September 1995 (Simmons 1995). Water retention modeling for the Ferrocyanide Safety Program has been completed. However, additional saltcake modeling continues for the Organic Safety Project.

### 3.5 CHEMICAL REACTION STUDIES

"The schedule for the program on study of the chemical properties and explosive behavior of the waste in these tanks is indefinite and does not reflect the urgent need for a comprehensive and definitive assessment of the probability of a violent chemical reaction. The study should be extended to other metallic compounds of ferrocyanide that are known or believed to be present in the tanks, so that conclusions can be generalized as to the range of temperature and other properties needed for a rapid chemical reaction with sodium nitrate."

Chemical reaction studies on ferrocyanide waste simulants have been conducted by Westinghouse Hanford Company, FAI, PNNL, and Los Alamos National Laboratory (LANL). All of the programs have been completed. Both Westinghouse Hanford Company and PNNL previously produced flowsheet simulant materials for testing and characterization. Earlier, FAI conducted adiabatic calorimetry and propagation tests on these flowsheet materials and on stoichiometric mixtures of pure sodium nickel ferrocyanide and sodium nitrate/nitrite. The test program at LANL evaluated the sensitivity of ferrocyanide reactions to shock, friction, and thermal aging (Cady 1993) and was completed in FY 1993.

### 3.5.1 Chemical Reaction Studies at Pacific Northwest National Laboratory

Chemical reaction studies at PNNL were completed in June 1996 using flowsheet simulant materials. Waste studies addressing DNFSB Recommendation 90-7.5 were conducted to determine the following: aging effects (hydrolysis and radiolysis) from more than 35 yr of storage in the tanks, reaction kinetics and properties of various mixtures of ferrocyanide with sodium nitrate and/or sodium nitrite, comparison of waste simulant and actual waste properties, and modeling calculations to predict the moisture-retaining capability of ferrocyanide waste in a typical tank system. All of these tasks are now complete.

• Status. All work is complete for this task and DNFSB Recommendation 90-7.5 is closed. See the final aging report for this task, PNNL-11211 (Lilga et al. 1996), for details.

### 3.5.2 Preparation and Characterization of Ferrocyanide Simulants

Earlier in the Ferrocyanide Safety Program, various ferrocyanide waste simulants were prepared and analyzed to determine their chemical reaction properties as a function of temperature and water present in the mixture. Other tests were conducted with pure sodium nickel ferrocyanide, a stoichiometric amount of oxidizer (sodium nitrate/nitrite), and water. The latter tests were conducted by FAI to define the margin of safety between the theoretical and experimental propagation limits for ferrocyanide. These tests were run in the FAI reactive systems screening tool. These and previous tests with simulants, along with analyses of actual tank waste samples, waste tank monitoring, and waste modeling, provide

information to characterize with a large degree of conservatism the safety concerns relating to the ferrocyanide sludge originally added to 18 of the Hanford Site waste tanks.

• Status. During the second quarter of FY 1996, a financial change request that deleted further chemical reactivity work at FAI on the Ferrocyanide Safety Issue was approved. Further work was not required because previous chemical reaction studies and results obtained by sampling nine of the ferrocyanide tanks show that the fuel value of ferrocyanide has degraded (aged) to levels too low to be of concern.

#### Milestone Status.

September 30, 1996. Complete FAI support for Ferrocyanide Safety Issue resolution and conclude chemical reactivity studies of chemical waste.
 This milestone was deleted by the financial change request approved last quarter.

### 3.6 EMERGENCY RESPONSE PLANNING

"The Board had recommended 'that an action plan be developed for the measures to be taken to neutralize the conditions that may be signaled by alarms.' Two types of measures are implied: actions to respond to unexpected degradation of a tank or its contents, and actions to be taken if an explosion were to occur. Your implementation plan stated that 'the current contingency plans ... will be reviewed and revised if needed.' We do not consider that this proposed implementation of the Board's recommendation is adequately responsive. It is recommended that a written action plan founded on demonstrated principles be prepared as soon as possible, that would respond to indications of onset of abnormal temperatures or other unusual conditions in a ferrocyanide-bearing tank, to counter any perceived growth in hazard. A separate emergency plan should be formulated and instituted, covering measures that would be taken in event of an explosion or other event leading to an airborne release of radioactive material from the tanks, and that would protect personnel both on and off the Hanford Site. The Board believes that even though it is considered that the probability is small that such an event will occur, prudence dictates that steps be taken at this time to prepare the means to mitigate the unacceptable results that could ensue."

The original Action Plan for Response to Abnormal Conditions in Hanford Radioactive Waste Tanks Containing Ferrocyanide (Cash and Thurman 1991) was prepared in response to DNFSB Recommendation 90-7.6. The plan describes the steps to be taken if a temperature-increase trend above the tank temperature baseline is measured in any of the ferrocyanide tanks. The document was revised to include the monitoring criteria and responses for abnormal levels of flammable and toxic gases, as well as the reporting requirements, if established criteria are exceeded (Cash and Thurman 1992). The second revision of the plan was released in June 1994 (Fowler 1994).

The Tank Farm Stabilization Plan For Emergency Response, WHC-SD-PRP-TI-001, Revision 0 (WHC 1991) was issued in March 1991. If a radioactive release from a ferrocyanide tank were to occur, it would be detected by one or more radiation monitoring systems. Significant airborne or ground surface releases that spread beyond the immediate tank or tank farm would be detected by the tank farm area radiation detectors. These monitoring systems are on all tank farms. An emergency involving an underground radioactive waste storage tank is a unique event with potentially serious consequences both on site and off site. The stabilization plan provides quick, preplanned actions that can be used to stabilize an emergency event at an underground radioactive waste storage tank.

All actions with respect to emergency planning, emergency event recognition, protective action recommendations, and emergency response procedures have been completed. Further revisions and occasional validation exercises will be accomplished as part of the normal Westinghouse Hanford Company and DOE emergency planning efforts. No further reporting on these issues is planned, and this part of DNFSB Recommendation 90-7.6 is considered complete and closed.

DOE considers this recommendation to be closed with the provisos that the abnormal-conditions response plan and emergency plans are reviewed periodically and revised and updated as required to incorporate any additional controls determined appropriate by the ongoing Waste Tank Safety Program investigations. The Action Plan for Response to Abnormal Conditions in Hanford Site Radioactive Waste Tanks Containing Ferrocyanide was updated and released in June 1994 (Fowler 1994); validation exercises for various waste tank accident scenarios are conducted periodically (exercises for the tank farms are conducted every 2 years).

• Status. As noted in previous reports, all of the planned milestones for this task have been completed.

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### 4.0 IMPLEMENTATION OF THE WYDEN AMENDMENT

The Wyden Amendment (Public Law 101-510, 1990) requires that:

"...the Secretary of Energy shall identify which single-shelled or double-shelled high-level nuclear waste tanks at the Hanford Nuclear Reservation, Richland, Washington, may have a serious potential for release of high-level waste due to uncontrolled increases of temperature or pressure. After completing such identification, the Secretary shall determine whether continuous monitoring is being carried out to detect a release or excessive temperature or pressure at each tank so identified. If such monitoring is not being carried out, as soon as practicable the Secretary shall install such monitoring, but only if a type of monitoring that does not itself increase the danger of a release can be installed."

### 4.1 THE WATCH LIST

In March 1989, using process knowledge, process records, transfer records, and log books, Westinghouse Hanford Company identified 22 Hanford Site tanks as potentially containing 1,000 g-mol (211 kg or 465 lb) or more of ferrocyanide as the Fe(CN)<sup>6</sup> anion (Nguyen 1989). To avert possible injury to personnel and damage to the facility or environment, strict controls were identified for these and other tanks with safety issues. These controls were described in *Operating Specifications for Watch List Tanks*, OSD-T-151-00030, Revision 0 (WHC 1990). Tanks identified by this document have been commonly referred to as Watch List tanks (see WHC 1996 for latest revision). Two additional ferrocyanide tanks were identified in January 1991 (Borsheim and Cash 1991), increasing the number of ferrocyanide tanks to 24.

In November 1990, the Wyden Amendment (Public Law 101-510, 1990) was enacted. This law required the identification of Hanford Site tanks that may have a serious potential for release of high-level waste. In February 1991, the 24 ferrocyanide tanks were among the tanks identified (Harmon 1991), and were included in the subsequent July 1991 report to Congress that responded to the Wyden Amendment (Watkins 1991). However, reexamination of the historical records indicated that 6 of the 24 tanks did not contain the requisite 1,000 g-mol of ferrocyanide (Borsheim and Simpson 1991). Therefore, these six tanks should not have been included on the original Watch List or identified in the response to the Wyden Amendment. The six tanks were subsequently removed from the Watch List (Anttonen 1993, Sheridan 1994b). In addition, these tanks did not contain greater than 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub>, which also should have excluded them from the Watch List.

As part of the overall safety screening module being conducted by Westinghouse Hanford Company Tank Waste Remediation System, all of the Hanford Site SSTs will be core-sampled and characterized. All 18 tanks formerly on the Ferrocyanide Watch List have been removed (Kinzer 1996a, 1996b) and no further work related to ferrocyanide is planned.

Work conducted since 1991 on ferrocyanide reactions has resulted in a change in the criterion that would now be applied to place tanks on the Watch List if ferrocyanide was still a safety issue. The 1,000 g-mol inventory criterion previously used would be replaced with a fuel concentration criterion of 115 cal/g of dry sample (this is an energy equivalent to a concentration of 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub> in the waste). This criterion was used as the basis for removing the 18 tanks from the Watch List because this fuel concentration criterion more accurately reflected the risk associated with the former ferrocyanide tanks. Tanks with concentrations less than an energy equivalent of 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub> cannot support a propagating reaction, and are categorized as safe. Detailed rationale for the 115 cal/g of dry fuel concentration criterion is presented in Ferrocyanide Safety Program: Safety Criteria for Ferrocyanide Watch list Tanks, WHC-EP-0691 (Postma et al. 1994).

Core sampling and characterization efforts were used to determine the ferrocyanide concentration for those tanks that bounded the ferrocyanide aging process (see Chapter 2 and Section 3.4). Because the bounding tanks showed insufficient ferrocyanide fuel values (much less than 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub>; i.e., the fuel value at the maximum concentration was less than 115 cal/g), a request was forwarded to DOE for concurrence to remove all the ferrocyanide tanks from the Watch List.

Because some sample bias and analytical error are unavoidable, confidence intervals were established to specify when it was appropriate to conclude that a ferrocyanide tank contained concentrations less than an energy equivalent of 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub>. An 80% confidence interval was chosen for tanks with a fuel concentration of 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub>. That is, if 5 ferrocyanide tanks contain exactly an energy equivalent of 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub>, statistically, 4 tanks would remain on the Watch List and 1 tank would be removed. The possibility of removing a ferrocyanide tank from the Watch List decreases substantially as the fuel concentration increases. The confidence intervals increase to 95% and 99% at Na<sub>2</sub>NiFe(CN)<sub>6</sub> concentrations of 12 wt% and 15 wt%, respectively. Detailed discussions on how sample bias and analytical error were factored into determining the actual fuel concentrations in a ferrocyanide tank are given in the Ferrocyanide DQO (Meacham et al. 1995).

- Planned Work To Complete Program. DOE has approved removal of all 18 ferrocyanide tanks from the Watch List (Kinzer 1996a, 1996b) and the DNFSB has closed Recommendation 90-7. Documentation is now being updated to show this change and the required final action to approve the change to the ISB will be transmitted to DOE-RL during the first quarter of FY 1997.
- Milestones.
  - January 31, 1996. Westinghouse Hanford Company issues documentation supporting safety issue resolution for the four C-Farm tanks and recommends their removal from the Wyden Amendment Watch List (Public Law 101-510, 1990). The Assessment of the Potential for Ferrocyanide Propagating Reaction Accidents, WHC-SD-WM-SARR-038,

Revision 0, was transmitted to DOE-RL as scheduled (Bacon 1996a, Grigsby et al. 1996a). All four C-Farm tanks were sampled earlier and data interpretation reports (TCRs) were completed for these tanks.

The SARR-038 report was revised to incorporate informal comments received from DOE-RL and reissued with the same title as WHC-SD-WM-SARR-038, Revision 0A, (Grigsby et al. 1996b). The revised report was transmitted to DOE-RL on March 19, 1996, with a letter requesting that the four C-Farm tanks be removed from the Watch List (Bacon 1996b). The report was subsequently forwarded to the DNFSB Staff on April 10, 1996 (Trine 1996).

- July 31, 1996. Westinghouse Hanford Company receives DOE approval to remove the four C-Farm ferrocyanide tanks from the Watch List. On June 25, 1996, DOE-RL approved removal of these tanks from the Watch List.
- March 31, 1997. Westinghouse Hanford Company prepares documentation to support removal of the last 14 ferrocyanide tanks from the Watch List and recommends final resolution of the Ferrocyanide Safety Issue. This milestone was accelerated to July 3, 1996, and the final safety document, WHC-SD-WM-SARR-038, Revision 1, (Meacham et al. 1996) was transmitted to DOE-RL on July 3, 1996 (Bacon 1996c).
- September 30, 1997. Westinghouse Hanford Company receives DOE approval to remove the remaining 14 ferrocyanide tanks from the Watch List and to declare the Ferrocyanide Safety Issue resolved. DOE approved removal of the 14 remaining tanks on September 4, 1996 (Kinzer 1996b). The DNFSB closed Recommendation 90-7 on September 4, 1996 (Conway 1996) and DOE-HQ has prepared a letter stating that the Ferrocyanide Safety Issue is resolved. This letter, when approved, will complete the Ferrocyanide Safety Program.

### 4.2 TEMPERATURE MONITORING

The installation of temperature monitoring capabilities is discussed in Sections 3.1.1 and 3.2. Installation of instrument trees and continuous temperature monitoring are considered prudent waste management practices. Therefore, new instrument trees were installed in the former ferrocyanide tanks, even though the ferrocyanide waste had aged and little or no fuel value remains. Instrument trees have been installed in all former ferrocyanide tanks and are continuously monitored by TMACS.

• Status. This task is complete.

#### 4.3 PRESSURE MONITORING

The former ferrocyanide tanks were initially identified as having "a serious potential for release" and were placed on the Watch List because insufficient data were available on the probability for ferrocyanide-nitrate/nitrite reactions. Pressure monitoring instrumentation is not presently installed on the former ferrocyanide tanks. It would take several years to install pressure monitoring instrumentation because of the capital project time cycle. Ferrocyanide waste has probably degraded (aged) significantly, and all of the tanks may now contain less than the 8 wt% Na<sub>2</sub>NiFe(CN)<sub>6</sub> fuel concentration specified for the safe category (Postma et al. 1994a). This eliminates the need for continuous pressure monitoring for offgases from a ferrocyanide reaction.

The rationale for not installing pressure monitors in the former ferrocyanide tanks was prepared and submitted to DOE in July 1994 (Payne 1994a). Low gas generation rates (Fowler and Graves 1994) and the low potential for exothermic ferrocyanide reactions (Postma et al. 1994a) indicated that continuous pressure monitoring is not warranted.

• Planned Work For Subsequent Months. No additional work is required in this area, because DOE has concurred that pressure monitoring is not required, as stated in the revised Ferrocyanide Safety Issue Program Plan (O'Leary 1994), and there are no longer any ferrocyanide tanks on the Watch List.

### 5.0 PROGRAM SCHEDULES AND MILESTONES

Schedules are presented in this chapter (see Figure 5-1). The schedules review milestones for FY 1994 through the expected end of the program in FY 1997, as described in the Ferrocyanide Safety Program Plan (DOE 1994b). The sequence and anticipated completion dates of the major milestones leading to resolution of the Ferrocyanide Safety Issue are presented. Closure of DNFSB 90-7 recommendations are indicated on the schedule as diamonds, and completion of interim milestones is indicated as triangles. The schedule status is shown through September 30, 1996. Note that DOE approval has been received to remove all 18 ferrocyanide tanks from the Watch List (Kinzer 1996a, 1996b) and that the DNFSB has closed Recommendation 90-7 (Conway 1996).

Figure 5-1.

Ferrocyanide Waste

Tank Safety Schedule. (Sheet 1 of

<sup>∇</sup> Interim Milestones

Closure of DNFSB Milestones

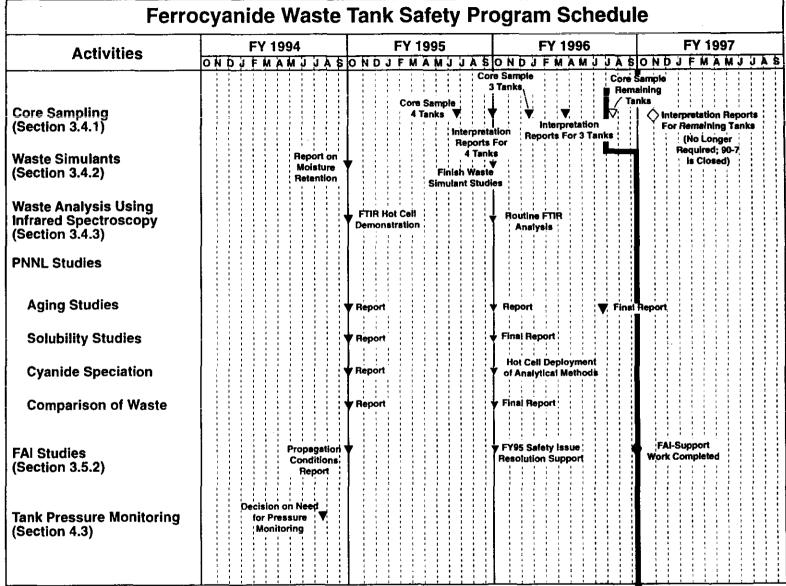
Figure 5-1.

Ferrocyanide Waste

Tank Safety Schedule.

(Sheet 2

of



<sup>∇</sup> Interim Milestones

29504077.2 Rev. Date 10/14/96

September 30, 1996

Closure of DNFSB Milestones

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FERROCYANIDE TANK INFORMATION SUMMARY

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Table A-1. Summary of Contents and Status of 18 Former Ferrocyanide Tanks.<sup>a</sup>

Tank	Total waste volume (1,000 L)	FeCN <sup>b</sup> (1,000 g-mol)	Heat load (kW) <sup>c</sup>	Maxii tem (°C)	ıp.	Riser No.	Status of tanks <sup>d</sup>
BY-103	1510	66	1.6	26 27°	79 80	1 5	NS; AL
BY-104	1540	83	3.3 <sup>f</sup>	50° 44	122 110	1 10B	IS; Sound
BY-105	1900	36	4.9 <sup>f</sup>	45 44	113 111	1 10C	NS; AL
BY-106	2430	70	4.7 <sup>f</sup>	50	123	1	NS; AL
BY-107	1010	42	2.6	36 37°	96 98	1 5	IS; AL
BY-108	863	58	2.7	42° 42	107 107	3 8	IS; AL
BY-110	1510	71	3.3 <sup>f</sup>	46 40°	115 105	1 10A	IS; Sound
BY-111	1690	6	2.1 <sup>f</sup>	30 <sup>g</sup> 28 <sup>c</sup>	87 83	1 (LOW) 14	IS; Sound
BY-112	1100	2	2.4 <sup>r</sup>	31° 308	89 85	2 15 (LOW)	IS; Sound
C-108	250	25	2.9 <sup>f</sup>	26° 25	78 77	1 5	IS; Sound
C-109	250	6.8 <sup>h</sup>	3.0 <sup>f</sup>	27° 27	80 80	3 8	IS; Sound
C-111	216	33	2.5 <sup>f</sup>	26 25°	78 77	5 6	IS; AL
C-112	394	11.5h	3.3 <sup>f</sup>	29 28 <sup>e</sup>	83 83	1 8	IS; Sound
T-107	681	5	1.2 <sup>f</sup>	20 19°	68 67	4 5	NS; AL
TX-118	1310	<3	1.4	25° 23	77 74	1 3	IS; Sound
TY-101	447	23	1.1 <sup>f</sup>	19 <sup>c</sup> 19	65 67	3 4	IS; AL

Table A-1. Summary of Contents and Status of 18 Former Ferrocyanide Tanks.<sup>a</sup>

Tank	Total waste volume (1,000 L)	FeCN <sup>b</sup> (1,000 g-mol)	Heat load (kW)°	Maximum temp. (°C) (°F)	Riser No.	Status of tanks <sup>d</sup>
TY-103	613	28	1.5	21 70 22° 71	4 7	IS; AL
TY-104	174	12	0.9	19° 67 19 66	3 4	IS; AL

### Notes:

- <sup>a</sup> Reflects removal of four ferrocyanide tanks from the Watch List in July 1993 and two additional tanks in October 1994. Note: all remaining tanks were removed from the Watch List in June 1996 (Kinzer 1996a) and in September 1996 (Kinzer 1996b). Tank information as of September 1996; temperatures are the highest recorded during the quarter ending September 30, 1996.
- <sup>b</sup> Original tank inventories (Borsheim and Simpson 1991).
- <sup>c</sup> Heat load values from Table 7-1 in Crowe et al. (1993).
- <sup>d</sup> IS Interim-Stabilized Tank; NS Not Stabilized; AL Assumed Leaker Tank; Sound Non-Leaking Tank.
- <sup>e</sup> Readings from new instrument trees; tank 241-BY-105 already had two trees.
- Data from Crowe et al. 1995.
- <sup>g</sup> Temperature reading from a single thermocouple in the LOW.
- <sup>h</sup> Calculated as ferrocyanide [Fe(CN)<sub>6</sub><sup>4</sup>] based on the total cyanide values reported in Simpson et al. (1993a, 1993b).

Tank	Date	Flamm.	Organic	NH,	NH,	HCN	NO+NO <sub>2</sub>	TNMOC	H <sub>2</sub>	N <sub>2</sub> O	CO	CO2	Water
	Sampled	(% LEL)b	Vapor	(ppmv)d	(ppmv)e	(ppmv)d	(ppmv) <sup>d</sup>	(mg/m³) <sup>f</sup>	(ppmv)1	(ppmv)*	(ppmv)#	(ppmv)*	Content
	(Type)"		(ppmv) <sup>c</sup>										%RH (°C) <sup>h</sup>
BY-103	05/05/94 (2)	<1	1.2	25	30.7	<0.005 <sup>i</sup>	<0.3	5.2	21.4	49.2	<1		
	11/01/94 (3)	ر			26		< 0.2		<99	16.5	< 12	126	49 (25.5°C)
BY-104	04/22/94 (2)	<1	26	200	285	< 0.005 <sup>1</sup>	< 0.3	56	204	305	<1		
	06/24/94 (3)				248		<0.4	61	295	201	1	10.5	58 (26 °C)
BY-105	05/09/94 (2)	<1	4.9	40	57	< 0.005	< 0.1	17.8	85	122	0.5		-
	07/07/94 (3)				43		< 0.2	12.7	48	50	0.4	94	61 (26 °C)
BY-106	05/04/94 (2)	<1	5.7	60	87	<0.01	<0.2	6.3	46	94			
	07/08/94 (3)				74		< 0.2	9.9	46	71	0.5	47.6	57 (27 °C)
BY-107	03/25/94 (2)	3 - 4	67	97				173	692	802	< 5		
	10/26/94 (3)				972		< 0.2	150	267	621	<20	94	36 (33.1°C)
BY-108	03/28/94 (2)	1	97	700			<0.5	594	644	757	<5		
	10/27/94 (3)	5	71.8		1040		< 0.1	510	400	641	<76	224	56 (25.7°C)
	1/23/96 (3)	0		700	801			222	361	470	<4	12	62 (25.0°C)
	3/28/96 (3)	0	3.3		822			243	352	505	<4	18	64 (24.4°C)
	9/10/96 (3)	7	68		953	<1		153	585	827	<4	<3	51 (28.3°C)
BY-110	09/27/92 (1)	<1	350	612		<2	< 0.5						
	11/11/94 (3)				401		< 0.2	29	<160	103	<76	229	31 (27 °C)
BY-111	05/11/94 (2)	<1	8.9	60							<b>₹1</b>		
	11/16/94 (3)			<u> </u>	59		< 0.2	9.6	67	99	<1	219	27 (27 °C)
BY-112	03/26/93 (1)	<1	5.9	10	***	<2	< 0.5	-					
	11/18/94 (3)				63		< 0.2	5.8	<94	40	<12	121	53 (23.3°C)
C-108	07/23/93 (**)	<1	1.2	<2		<2	< 0.5	-	••		1		
	07/07/94 (2)					<0.00021		<0.4			<u> </u>		
	08/05/94 (3)	<u> </u>			2.7		< 0.3	<1.4	15.3	344	0.1	16.3	76 (25 °C)
C-109	06/23/94 (2)	<1	1	4						260			
	08/09/94 (3)				10.1		<0.6	0.65		369	0.4	3	79 (27 °C)
C-111	08/10/93 (**)		< 0.2	<2		< 0.04 <sup>k</sup>	< 0.5	<0.3	16	39	0.1		
	06/20/94 (2)	<1	< 0.2	<2	0.1	<0.01	< 0.2	0.18			<del></del> ,	100	
	09/13/94 (3)				5.6		≤0.7	<0.6	12.4	99	0.1	198	86 (27 °C)
C-112	06/24/94 (2)	<1	< 0.2	4								100	
	08/11/94 (3)	1			22.7		< 0.7	3.4	204	544	0.9	102	82 (28 °C)

Table A-2. Ferrocyanide Tank Vapor Sampling Summary. (2 Sheets)

			Tuoic 71	2. 10.	rrocyan		vapor bu	inpinig of	anning y .	(2 511001	<del></del>		
Tank	Date Sampled (Type) <sup>a</sup>	Flamm. (% LEL) <sup>b</sup>	Organic Vapor (ppmv) <sup>c</sup>	NH <sub>3</sub> (ppmv) <sup>d</sup>	NH <sub>3</sub> (ppmv) <sup>e</sup>	HCN (ppmv) <sup>d</sup>	NO+NO <sub>2</sub> (ppmv) <sup>d</sup>	TNMOC (mg/m³) <sup>f</sup>	H <sub>2</sub> (ppmv) <sup>2</sup>	N <sub>2</sub> O (ppmv) <sup>2</sup>	(ppmv) <sup>s</sup>	CO <sub>2</sub> (ppmv) <sup>g</sup>	Water Content %RH (°C) <sup>h</sup>
T-107	10/22/92 (1) 01/18/95 (3)	<1 	24 	203	 125	<2 	<0.5 <0.1	 1.4	 <94	42	 <12	 75	 82 (17.2°C)
TX-118	07/28/93 (**) 09/07/94 (2) 12/16/94 (3)	<1 <1 	0.3 7.8 	10 28 	  33	<2 <0.02 	<0.5 <0.5 	9.3 	 97 <94	 17 29	2.5 <12	 54 98	 42 (21.5°C)
TY-101	08/04/94 (2) 04/06/95 (3)	<1 	4	12 	16 16	<0.01 	<0.2 <0.2	1.0	 <93	 98	 <12	 83	 77 (15.6°C)
TY-103	08/04/94 (2) 04/11/95 (3)	<1 	5	30 	31 49	<0.01	<0.1 <0.2	 60	 <93	 159	 <12	121	 85 (15.9°C)
TY-104	08/05/94 (2) 04/27/95 (3)	<1 	2.5	24	50 61	 	<0.2 ≤0.2	3	 <49	98	<23	 <23	 88 (15.6°C)

Table A-2. Ferrocyanide Tank Vapor Sampling Summary. (2 Sheets)

\* Sample Type:

\*\*Vapor samples taken from in-tank, non-heated tubes using a vapor sampling cart (SUMMA™ only - no NH₃).

- Monitoring performed by Industrial Hygiene technicians using three varying length, non-heated sampling tubes into the tank headspace to evaluate for flammability and toxic vapors; this method is no longer used.
- 2 In Situ Sampling (ISS) Sampling is performed by lowering special sorbent traps into the tank headspace that are connected topside to a portable handcart.
- 3 Sampling involves the mobile vapor sampling system, heated transfer lines, and installation of a water-heated sampling probe into the tank headspace.
- <sup>b</sup> Measured using a combustible gas meter; LEL = Lower Explosive Limit.
- <sup>c</sup> Measured using an Organic Vapor Monitor (OVM). OVM readings are affected by ammonia; OVM ammonia response is approximately 13:1, so that 13 ppmv of ammonia is indicated as 1 ppmv of organic vapors (ppmv = parts per million by volume).
- <sup>d</sup> For Type 1 sampling only; value is measured using colorimetric (Dräger™) tubes (values are estimated, and not quantitative).
- Analyses of ammonia sorbent trap samples.
- Total non-methane organic compound (TNMOC) concentrations measured for SUMMA<sup>TM</sup> canister samples.
- \* Analyses of SUMMA<sup>TM</sup> canister samples from Type \*\*, and 3 sampling methods.
- h % RH is the percent relative humidity calculated from measured headspace water content (mg/L), temperature and atmospheric pressure. Temperature of headspace gas in °C is listed in parentheses.
- HCN determinations obtained in selected tanks using a special sorbent trap; values shown are below detection limit of the measurement technique.
- i Data not yet available or not obtained by this type of sampling.
- <sup>1</sup> This HCN number was < 0.04 parts per billion vapor as determined by a special sodium hydroxide bubbler.

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